

# Non-Fermi-liquid ferromagnetic Kondo system CeRuSi<sub>2</sub>

V.N. Nikiforov<sup>1</sup>, M. Baran<sup>2</sup>, A. Jedjenchak<sup>2</sup>, V.Yu. Irkhin<sup>3\*</sup>

<sup>1</sup>*Department of Physics, Moscow State University, 119899 Moscow, Russia*

<sup>2</sup>*Institute of Physics, Polish Academy of Science, Lutnikow, Warsaw, Poland and*

<sup>3</sup>*Institute of Metal Physics, 620990 Ekaterinburg, Russia*

The structural, electronic and magnetic properties of the Kondo-lattice system CeRuSi<sub>2</sub> are experimentally investigated and analyzed in the series of other ternary cerium compounds. This system is shown to be an excellent model system demonstrating coexistence of the Kondo effect and anomalous ferromagnetism with a small magnetic moment which is confirmed by magnetic and  $\mu$ SR measurements. Data on specific heat, resistivity and Seebeck coefficient are presented. Being deduced from the resistivity and specific heat data, a non-Fermi-liquid behavior is observed at low temperatures, which is unusual for a ferromagnetic Kondo system. A comparison with other ferromagnetic Kondo lattices is performed.

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## I. INTRODUCTION

The cerium based ternary intermetallic compounds of CeTX<sub>2</sub> type (with T being transition metal and X semimetallic element like Si, Ge and Sn) remain a subject of considerable interest because of their unusual ground state properties observed in many compounds of this family. For example, we have heavy-fermion behavior with a large electronic specific heat ( $\gamma = 1.7$  J/(mol K<sup>2</sup>) at 1.25 K) in CePtSi<sub>2</sub> [1], valence fluctuation behavior in CeRhSi<sub>2</sub> [3] and in CeNiSi<sub>2</sub> [2]. However, the CeTX<sub>2</sub> series has not been studied completely yet. In the present work we discuss in detail the properties of the system CeRuSi<sub>2</sub> which has a ferromagnetic ground state [4].

It was traditionally believed for many years that the competition of the intersite RKKY exchange interaction and the Kondo effect should result in the formation of either the usual magnetic ordering with large atomic magnetic moments (as in elemental rare-earth metals) or the non-magnetic Kondo state with suppressed magnetic moments [5]. However, more recent experimental investigations have convincingly demonstrated that magnetic ordering and pronounced spin fluctuations are widely spread among heavy-fermion systems and other anomalous 4f- and 5f-compounds, which are treated usually as concentrated Kondo systems (see [6]).

The class of "Kondo" magnets is characterized by (i) the logarithmic temperature dependence of resistivity typical for Kondo systems at not too low temperatures (ii) reduced value of the magnetic entropy at the ordering point, in comparison with the value  $R \ln(2J + 1)$  (which corresponds to usual magnets with localized moment  $J$ ) (iii) small ordered magnetic moment  $M_0$  (in comparison with the "high-temperature" moment  $\mu_{eff}$  determined from the Curie constant), which is reminiscent of weak itinerant magnets (iv) negative (even for ferromagnets) paramagnetic Curie temperature  $\theta$  which strongly ex-

ceeds in absolute value the magnetic ordering temperature (this behavior is due to large single-site Kondo contribution to the paramagnetic susceptibility).

There exist numerous examples of antiferromagnetic systems where "Kondo" anomalies in thermodynamic and transport properties coexist with magnetic ordering. At the same time, examples of Kondo ferromagnets are not numerous: CeNiSb, CePdSb, CeSi<sub>x</sub>, Sm<sub>3</sub>Sb<sub>4</sub>, NpAl<sub>2</sub> (the bibliography is given in Ref.[6]), CePt [7], CeRu<sub>2</sub>Ge<sub>2</sub> [8], CeAgSb<sub>2</sub> [10], CeRuPO [9], and some more compounds.

In fact, a number of factors make the physical picture in the most "Kondo" ferromagnets rather complicated. The systems like CeSi<sub>x</sub> [13] are described by spin-fluctuation (rather than Kondo) model. The Kondo systems YbNiSn [11] and UPdIn [12] possess antiferromagnetic ordering and demonstrate canted ferromagnetism only.

In our opinion, CeRuSi<sub>2</sub> is a rather "typical" anomalous rare-earth ferromagnetic system which exhibits the whole variety of peculiarities of the Kondo-lattice magnets. Moreover, it demonstrates the phenomenon of the non-Fermi-liquid behavior [14] in low-temperature thermodynamic and transport properties.

## II. THE CRYSTAL STRUCTURE

We have investigated a large number of CeTX<sub>2</sub>, CeT<sub>2</sub>X<sub>2</sub>, Ce<sub>2</sub>T<sub>3</sub>X<sub>5</sub> and CeTX<sub>3</sub> compounds (with T = Ru, Rh, and X = Si, Ge). The high-purity samples were provided by Physical and Chemical Analysis Laboratory from the Chemical Department of the Moscow State University (guided by Yu. Seropenin). According to their crystal symmetry, these compounds can be divided into following classes: CeRuSi<sub>2</sub> (space group P21/m); CeRu<sub>2</sub>Si<sub>2</sub>, CeRu<sub>2</sub>Ge<sub>2</sub>, CeRh<sub>2</sub>Si<sub>2</sub>, CeRh<sub>2</sub>Ge<sub>2</sub> (I4/mmm); CeRuSi<sub>3</sub>, CeRuGe<sub>3</sub>, CeRhSi<sub>3</sub> (I4mm); Ce<sub>2</sub>Ru<sub>3</sub>Ge<sub>5</sub>, Ce<sub>2</sub>Rh<sub>3</sub>Si<sub>5</sub> (Ibam).

In the present work we focus on the most interesting compound CeRuSi<sub>2</sub>, although give in Sect. 4 a compari-

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\*Electronic address: Valentin.Irkhin@imp.uran.ru

son with resistivity data for other systems mentioned.

The polycrystalline samples of  $\text{CeRuSi}_2$  were synthesized by melting the starting mixture in an arc furnace in an argon atmosphere followed by annealing at 870K for 600h. The purity of the component metals was better than 99.9%. Our investigation of  $\text{CeRuSi}_2$  [4] has shown that this compound crystallizes in a structure different from the  $\text{CeNiSi}_2$  type which is common for other compounds of the  $\text{CeTX}_2$  series. The X-ray investigations in Moscow (Yu. Seropegin) and Lvov (O. Bodak) demonstrated that this compound has the  $\text{NdRuSi}_2$ -type low-symmetrical monoclinic crystal structure. The sample material was examined by X-ray analysis and identified as a single  $\text{CeRuSi}_2$  phase (the calculated X-ray density was approximately  $9.485 \text{ g/cm}^3$ ).

The  $\text{NdRuSi}_2$  structure is a distortion derivative of the orthorhombic  $\text{CeNiSi}_2$ -type structure characteristic for other  $\text{CeTX}_2$  compounds. The unit cell parameters for  $\text{CeRuSi}_2$  are:  $a = 4.478(1)\text{\AA}$ ,  $b = 4.093(1)\text{\AA}$ ,  $c = 8.302(5)\text{\AA}$ , the angle beta being equal  $102.53(3)^\circ$ . The atomic coordinates are:

Ce	2(e)	x/a = 0.4130(2)	y/b = 1/4	z/c = 0.79904(9)
Ru	2(e)	x/a = 0.1179(2)	y/b = 1/4	z/c = 0.3869(2)
Si1	2(e)	x/a = 0.0364(9)	y/b = 1/4	z/c = 0.0907(5)
Si2	2(e)	x/a = 0.6657(9)	y/b = 1/4	z/c = 0.4913(3)

The  $\text{CeRuSi}_2$  sample used in further measurements was of the weight 351.57 mg and of disk shape with the diameter of approximately 9 mm and thickness 1 mm.

### III. MAGNETIC PROPERTIES

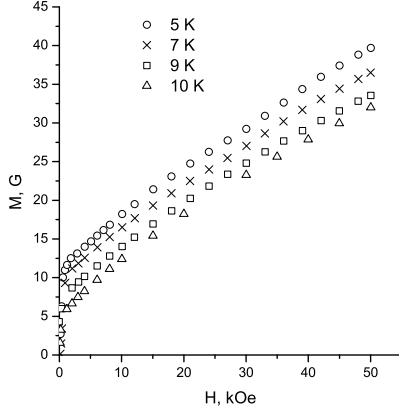


FIG. 1: The field dependences of magnetization at different temperatures

The magnetization was measured by a SQUID magnetometer of Quantum Design MPMS-5 in the broad interval of magnetic fields ( $H < 50 \text{ kOe}$ ) and temperatures

( $5 < T < 250 \text{ K}$ ). These data give evidence for the coexistence of the Kondo effect and magnetism in  $\text{CeRuSi}_2$ .

In the paramagnetic region (investigated up to 260 K) the simple Curie-Weiss law is not fulfilled (see Fig. 2). However, analyzing experimental data we were able to describe them by taking into account the Van Vleck susceptibility which is independent of temperature. Fitting  $M/H = M_0/H + C_{eff}/(T - \theta)$  to the experimental data, it was possible to get  $M_0/H = 3.710^{-6} \text{ emu/g Oe}$ ,  $C_{eff} = 1.22 \cdot 10^{-4} \text{ emu/g Oe}$  and  $\theta$  about  $-40 \text{ K}$ . The value of  $M_0/H$  is about 50% of  $M/H$  for 260K. This large Van Vleck contribution to the susceptibility is probably connected with low symmetry positions of Ce ions.

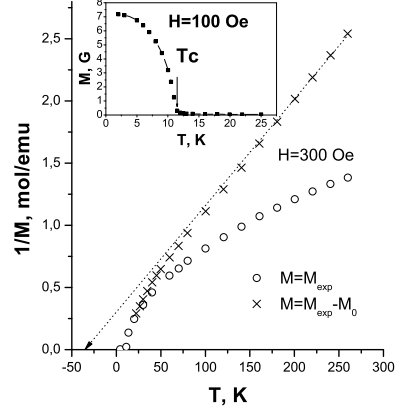


FIG. 2: The extraction of the Van Vleck contribution The inset shows the temperature dependence of magnetization at low temperatures

Thus the temperature dependence of magnetic susceptibility at temperatures above 60 K can be described by the Curie-Weiss law with the Curie constant  $C_{eff}$ , the corresponding value of the effective magnetic moment  $\mu_{eff}$  being equal  $1.7\mu_B$  which is somewhat smaller in comparison to that of the free  $\text{Ce}^{3+}$  ion. The reduced value of the effective magnetic moment  $\mu_{eff}$ , calculated from the linear  $\chi^{-1}(T)$  dependence, can be partly attributed to a moderate renormalization of the 4f shell moments due to hybridization with conduction electrons. At the same time, magnetic fluctuations in the presence of the Jondo effect can play a role.

The paramagnetic Curie temperature  $\theta$  is negative. As discussed in the Introduction, the latter fact is typical for Kondo lattices (even ferromagnetic ones) since  $\theta$  is determined by on-site Kondo screening rather than by intersite exchange interactions,  $\chi(T = 0) \sim 1/T_K$ .

Fig. 1 demonstrates the field dependence of magnetization  $M(H)$  at four temperatures in the paramagnetic region. The  $M(H)$  curve in the reversible range of fields (from 5 to 50kOe) are close to linear and are characterized by a relatively significant slope ( $dM/dH$  is about  $5 \cdot 10^{-5} \text{ emu/g Oe}$ ) that depends slightly on temperature.

Saturation of the magnetic moment in strong fields is absent (up to 150 T, as measured by Arsamas equipment).

The dependence  $M(H)$  below the critical temperature  $T_c$  in the reversible range of fields is also characterized by relatively strong slope  $dM/dH$  (with the same value  $5 \cdot 10^{-5}$  emu/g Oe), slightly depending on temperature in the range of 5-10 K).

The inset in Fig. 2 shows the temperature dependences of magnetization measured in the field  $H = 100$  Oe. A sharp upturn of  $M(T)$  (as well as in the  $\chi(T)$  curve) below  $T_1$  and a tendency to saturation at lowest temperature were detected. Thus the shape of the  $M(T)$  and  $M(H)$  corresponds to the ferromagnet with the transition temperature  $T_1 = T_c$ .

The dependence  $M(0) - M(T)$  below  $T_c$  is close to  $T^{4/3}$ . Such a dependence is obtained in spin-fluctuation theories [15] and is typical for weak itinerant ferromagnets where the spin-wave picture works at very low  $T$  only (as discussed in the Introduction, these systems have a number of similar features with the Kondo magnets).

We have found hysteresis loops for 5, 6, 7, 8, 9 and 10 K (see inset in Fig. 3). From these loops we have estimated the coercive field  $H_c$  as one half of loop width.

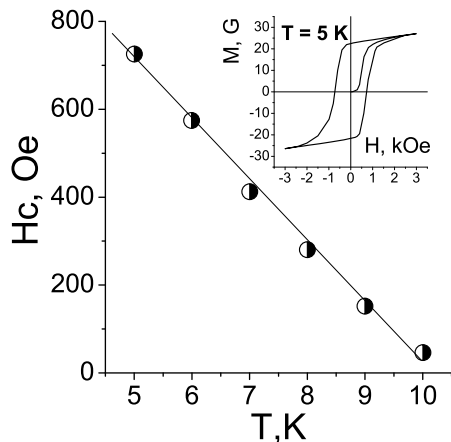


FIG. 3: The temperature dependence of the coercive field. The inset shows hysteresis loop for  $T = 5$  K

Fig. 3 shows the temperature dependence of  $H_c(T)$  as determined from the hysteresis loops at different temperatures.

Taking the  $T^{4/3}$  extrapolation of the  $M(T)$  most steep part (between 10.5 and 11 K) to  $x$ -axis we should get  $T_c = 11.7$  K. From the above  $M(H)$  data we have determined  $M_s$  values extrapolating linear dependence of  $M$  in the  $H = 20 - 50$  kOe range to  $H = 0$ .

The determination of the ferromagnetic transition temperature was also performed by the Belov-Arrott method. The field dependences of magnetization for  $T = 5-10$  K are presented in Fig. 4 as the plots  $M^2 = f(H/M)$ . Extrapolating  $M^2$  to the value  $H/M = 0$  enables one to obtain the value  $M_s^2$ , its temperature dependence yield-

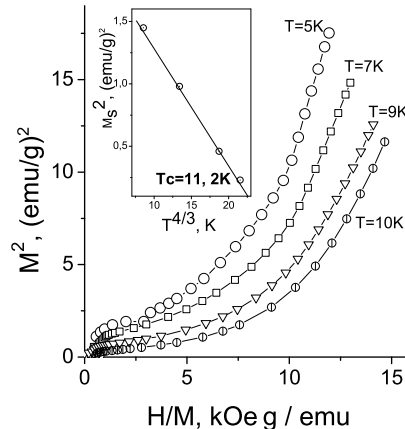


FIG. 4: Arrott plots of magnetization which determine the Curie temperature. Inset shows the determination of  $T_c$  from these plots

ing  $T_c = 11.2$  K. The plot of  $M_s^2(T)$  determining  $T_c$  is shown in inset of Fig. 4.

The relatively large slope in the reversible region of  $M$  and the linearity in the  $M(H)$  curves below the magnetic transition can be explained by the small value of the effective ground-state magnetic moment per Ce ion. The value  $M = 4$  emu/g obtained corresponds to  $\mu_s = 0.21\mu_B/\text{Ce ion}$ . This may give evidence for a complicated magnetic structure or, more likely, for a moment suppression by Kondo scattering which is responsible for the reduction of the  $\mu_{eff}$  value at low temperatures.

To probe low-temperature magnetism,  $\mu$ SR investigations were also performed on the muon channel of the LNP JINR phasotron using the spectrometer MUSPIN (some results were reported earlier [16]). Zero field (ZF), longitudinal field (LF) and transverse field (TF)  $\mu$ SR measurements have been carried out with a polycrystalline CeRuSi<sub>2</sub> sample. The temperature interval was  $4.2 \text{ K} < T < 300 \text{ K}$ , above and below the Curie temperature  $T_c$ . At all the temperatures, the muon spin polarization function  $P(t)$  has a non-oscillating form. Below  $T_c$  we found a hysteresis-like behavior of the polarization versus longitudinal magnetic field.

The lack of information about the muon site and type of magnetic ordering in the compound does not allow the precise determination of the value of the ordered moment. Nevertheless, taking into account the wide distribution of the magnetic field, we can estimate the value of the ordered magnetic moment about  $0.05 \mu_B$ .

ZF data show a sharp increase of the muon relaxation rate below the temperature  $T = 11.7$  K ( $0.42 \mu\text{s}^{-1}$  at  $T = 4.2$  K) justifying the phase transition to the magnetically ordered state. The results of LF measurements at  $T = 4.2$  K show that the magnetic fields on the muon  $B_\mu$ , produced by the cerium magnetic moments, are mainly static: the external longitudinal field of 150 Oe practi-

cally recovers the muon spin polarization. In the paramagnetic phase the polarization decay has an exponential form at the temperatures  $20 \text{ K} < T < 300 \text{ K}$ , although LF experiments at  $T = 20 \text{ K}$  clearly show a significant static contribution (about 75%). This situation can be discussed in the framework of a double relaxation model. The ferromagnetic type of magnetic ordering was proved by a hysteresis  $B - H$  behavior observed in TF-experiment.

#### IV. THERMODYNAMIC AND TRANSPORT PROPERTIES

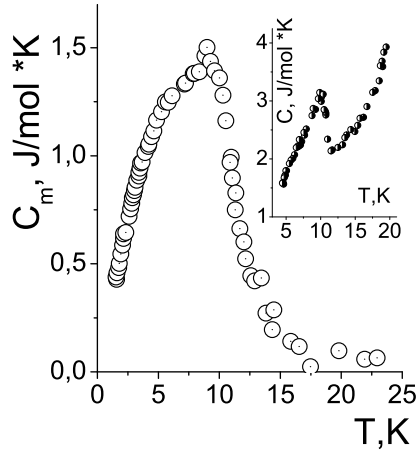


FIG. 5: The magnetic specific heat (after subtracting the  $T$ -linear and phonon contribution). The inset shows the temperature dependence of total specific heat

The specific heat anomaly at  $T_1$  (Fig. 5) confirms the presence of a magnetic transition. The entropy change near  $T_1$  as calculated by integrating the  $C(T)$  anomaly is relatively small:  $\Delta S = 2.7 \text{ J/K} = 0.6 R \ln 2$  (where  $R$  is the universal gaseous constant). The low value of  $\Delta S$  allows us to consider the 4f-free ion level of Ce to be split by a crystal electric field (CEF) with the doublet ( $J = 1/2$ ) as a ground state. The high temperature properties ( $T \gg T_1$ ) are therefore determined by the combined action of Kondo effect and CEF splitting.

The low temperature  $T$ -linear electronic specific heat was obtained by the standard method as shown in Fig. 6. The coefficient  $\gamma = 143 \text{ mJ/mol K}^2$  is markedly enhanced which means a moderately heavy-fermion behavior.

The fitting of specific heat in the region of low and extremely low temperatures demonstrates the logarithmic divergence  $C/T \text{ (J/mol K}^2\text{)} = (3.25 - 1.08 \log T) 10^{-4}$ . This means a non-Fermi-liquid (NFL) behavior. The temperature dependence can be also fitted by power-law dependences with small exponents (Fig. 7).

One of the ways to govern the NFL behavior is applying the external magnetic field [17]. Fig. 7 shows the

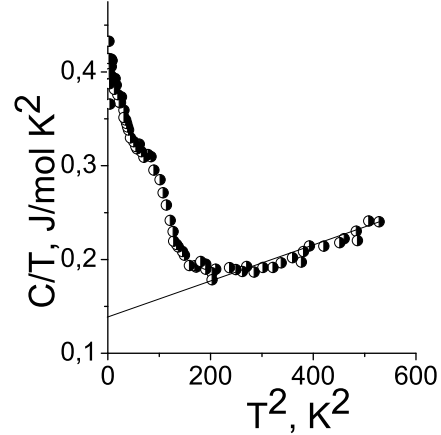


FIG. 6: The determination of  $T$ -linear electronic specific heat

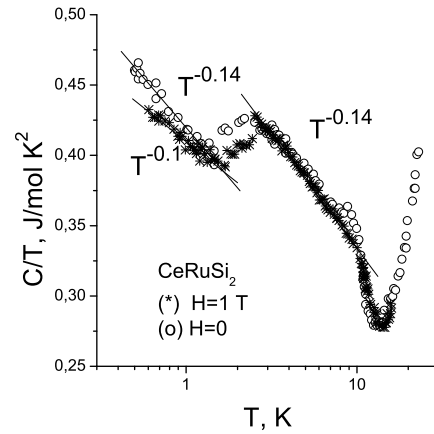


FIG. 7: The fit of temperature dependence of specific heat at ultralow temperatures in zero field and applied field  $H = 1 \text{ T}$

influence of magnetic field  $H = 1 \text{ T}$  which suppresses somewhat the corresponding  $C(T)$  anomaly (note that internal magnetic field in the ferromagnetic phase from  $\mu\text{SR}$  makes up 0.1 T only [16]). Unfortunately, we were not able to perform the measurements in more strong fields which are required to clarify details of NFL behavior. Therefore further investigations are of interest.

Specific heat has also a weak anomaly near 2 K which is also influenced by magnetic field and may be related to crystal field effects.

The temperature dependence of resistivity  $\rho(T)$  is shown in Fig. 8. This exhibits an anomaly at the temperature  $T_1 = 11.7 \text{ K}$ , a rapid decrease of  $\rho(T)$  below  $T_1$  being detected. Thus we can propose that the magnetic transition at 11.7 K suppresses the Kondo scattering and leads to a decrease of the low-temperature resistivity.

The second anomaly (a maximum) occurs at high temperatures. To extract this, we eliminated the phonon

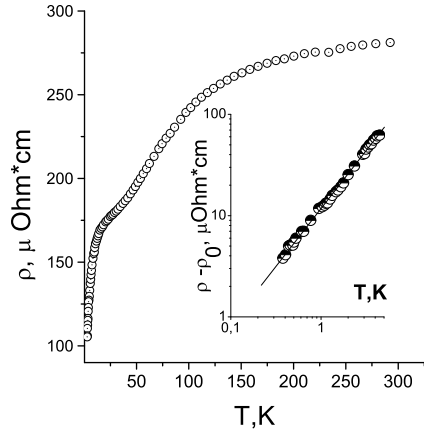


FIG. 8: The temperature dependence of resistivity in a wide temperature region. The inset shows the resistivity in the low temperature region

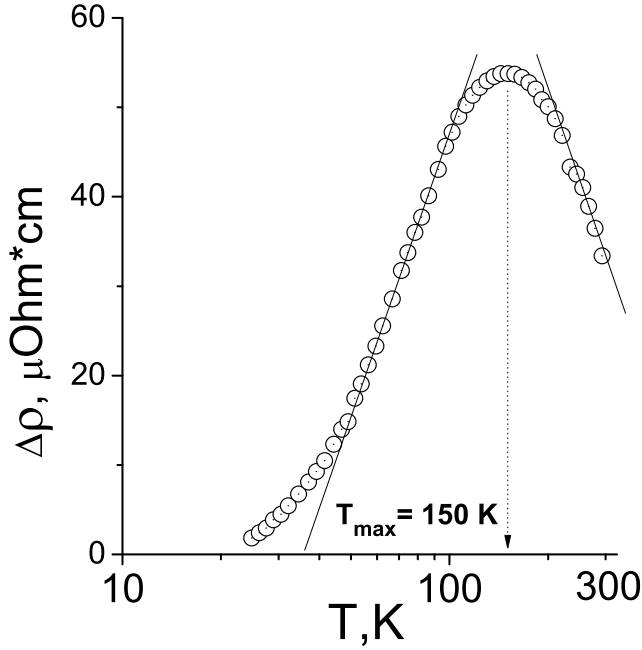


FIG. 9: The temperature dependence of resistivity at high temperatures after subtracting the phonon contribution

contribution by using the Debye model. The Debye temperature as determined from the  $T^3$ -contribution to specific heat (see Fig. 6) makes up  $\theta_D = 226$  K. After subtracting the corresponding Bloch-Grüneisen contribution we obtain the resistivity maximum at  $T_2 \simeq 150$  K (Fig. 9). Above  $T_2$  we may pick up a high-temperature logarithmic Kondo contribution to resistivity.

It should be noted that for the widely investigated system  $\text{CeRu}_2\text{Si}_2$  we have observed only one maximum ( $T_1 = 50$  K), and the system  $\text{Ce}_{15}\text{Ru}_{57}\text{Si}_{28}$  demonstrates a further shift of the maximum to higher temperatures

( $T_1 = 120$  K). On the other hand, for  $\text{CeRu}_2\text{Ge}_2$  we have observed two resistivity anomalies at  $T = 8$  K and 160 K. The first anomaly is accompanied by hysteresis and can be identified with a ferromagnetic transition, in agreement with the results of Ref.[8]. A magnetic transition was found also for  $\text{CeRuGe}_3$  at  $T = 7$  K. Following to the Sereni classification [18], the difference in magnetic behavior of various (binary and ternary) cerium compounds may be attributed to different CeCe distances.

In the low-temperature region, the resistivity of  $\text{CeRuSi}_2$  obeys a non-Fermi-liquid (NFL) law, namely,  $\rho(T) \propto T^\mu$  with  $\mu = 1.1 - 1.2$  rather than a square dependence (inset in Fig.8).

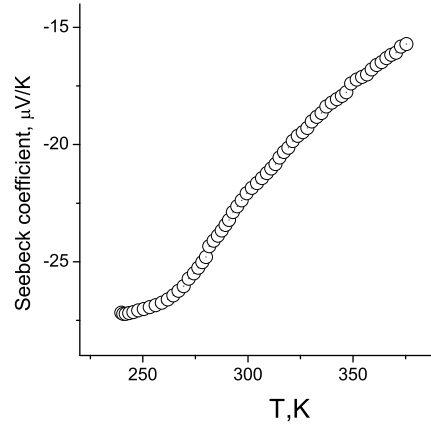


FIG. 10: The temperature dependence of thermoelectric power at high temperatures

The Seebeck coefficient measured at high temperatures (Fig.10) is rather high. Its temperature dependence seems to confirm the presence of the Kondo effect (cf. the discussion of thermoelectric power in the Kondo lattices in Ref. [19]). The corresponding figure of merit  $ZT$  has a maximum at  $T = 250$  K where it makes up about 0.6%.

## V. CONCLUSIONS AND DISCUSSION

To summarize, the results of the present paper and Ref.[4] allow us to conclude that  $\text{CeRuSi}_2$  belongs to a relatively rare family of compounds where the Kondo effect and a ferromagnetism coexist. The Kondo nature of ferromagnetism is confirmed by investigation of a wide range of electronic properties. The shape of  $M(H)$  and  $M(T)$  curves, observation of hysteresis loops together with lambda-peak in the temperature dependence of specific heat  $C(T)$  (see below) and the resistivity decrease are the characteristic features of a ferromagnetic transition at  $T_1 = 11.7$  K. An enhanced value of  $\gamma \simeq 140$  mJ/mol K<sup>2</sup>, reduced ground state moment, large absolute value of resistivity (being of hundreds  $\mu\text{Ohm cm}$ ) together with observation of a maximum in  $\rho(T)$  at

$T_2 \simeq 150$  K, are characteristic features of dense Kondo effect. Thus, CeRuSi<sub>2</sub> might be considered as a ferromagnetic Kondo lattice.

At the same time, formation of a complicated non-collinear magnetic structure cannot be excluded, similar to the situation in YbNiSn (canted ferromagnetism [11]). Neutron scattering experiments are required to investigate this issue. Being deduced from the resistivity and specific heat data at ultralow  $T$ , the non-Fermi-liquid behavior is observed in our ferromagnetic Kondo compound.

To explain the NFL behavior in rare-earth and actinide systems, a number of mechanisms were proposed [14]. Since many known NFL systems are disordered alloys, one considers the influence of disordering in the Kondo-lattice model [20], and the Griffiths singularity model [21].

In most Kondo magnets (see the Introduction) the NFL behavior can occur only on the boundary of magnetic instability, i.e. in the proximity of quantum critical point (quantum phase transition, QPT), which is governed by chemical composition or external pressure. In this connection, mechanisms connected with spin fluctuations [15], in particular treating the behavior near QPT in the clean limit [22] or with account of disordering [23].

An example is the URh<sub>1-x</sub>Ru<sub>x</sub>Ge alloy [24] where the critical concentration for the suppression of ferromagnetic order is  $x_{cr} = 0.38$ . The Curie temperature vanishes linearly with  $x$  and the ordered moment  $\mu_s$  is suppressed in a continuous way ( $\mu_s = 0.4\mu_B$  for  $x = 0$ ). At  $x_{cr}$ , the specific heat behaves as  $T \ln T$ , and the temperature exponent of the resistivity attains a minimal value

$\mu = 1.2$ . The total f-electron entropy obtained by integrating  $C_m/T$  equals  $0.48R \ln 2$  for  $x = 0$  and decreases to  $0.33R \ln 2$  at  $x_{cr}$ .

Recently, a picture somewhat similar to CeRuSi<sub>2</sub> has been reported for the ferromagnetic system URu<sub>2-x</sub>Re<sub>x</sub>Si<sub>2</sub> [25] which is characterized by a small ground state moment. In this system,  $C(T) = T \ln T$ ,  $\rho(T) \propto T^\mu$  with  $\mu = 1.2$  over more than a decade in temperature below 20 K for  $x = 0.6$ . The saturation moment, Curie temperature and electronic specific heat coefficient exhibit maxima for different Re concentration, reaching the values  $\mu_s = 0.44\mu_B/\text{U atom}$ ,  $T_c = 38$  K at  $x = 0.8$ , and  $\gamma = 160 \text{ mJ/mol K}^2$  at  $x = 0.6$ . The ferromagnetic order was confirmed by neutron scattering experiments for  $x = 0.8$  and NMR measurements for  $x = 0.4$ . However, corresponding magnetic-transition anomalies in specific heat and resistivity were not detected (unlike our investigations of CeRuSi<sub>2</sub>). The Griffiths-McCoy phase model [21] was discussed in Ref.[25] as an explanation for the coexistence of ferromagnetism and NFL properties since the Griffiths singularities give rise to the NFL power-law behavior. In a clean system, spin-fluctuation mechanisms of NFL behavior, e.g., owing to peculiarities of magnetic ordering and spin dynamics in the Kondo lattices [26], are more probable.

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